DISINTEGRATION OF BORON BY SLOW NEUTRONS

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(Plate V)

ABSTRACT. The disintegration of boron by slow neutrons has been investigated using an ionisation chamber filled with boron trichloride in conjunction with a linear amplifier and loop oscillograph.

The various factors which can affect the size of oscillograph deflections and thereby introduce a distortion in the shape of the distribution curve have been discussed.

The experimental evidence points to a reaction energy of about 2.5 eMV. It appears that the transition to the ground state of $_3Li^7$ is forbidden, and that the reaction leads to an excitation state in the $_3Li^7$ nucleus, with different angular momentum for which the transition is allowed.

INTRODUCTION

The nuclear transformation of Boron by slow neutrons has been the subject of several investigations. With slow neutrons, three different particle-disintegration processes are energetically possible. These are :

(1) $_5B^{10}((n, \alpha)_3Li^7$

(2) $_5B^{10}(n, p)_4Be^{10}$

and (3) $_5B^{10}(n, 2\alpha)_1H^3$

From the known mass-data occurring on both sides, we get Q = 2.98 eMV and Q = 0.41 cMV respectively for the first two reactions. The third involves a double process and takes place only with fast neutrons. For the investigation with particle emission, one can proceed in two different ways: Firstly, one can measure, in a cloud chamber, the sum of the ranges $R_\alpha + RLi$, from the photographs of the tracks and ascertain the energy-balance from the range-energy relation of α-particles, given by Bethe (1937), or secondly, one can directly estimate the energy release $E_\alpha + ELi$ with an ionisation chamber and proportional amplifier or Hoffmann electrometer. The energy of incident slow neutrons may be taken to be zero and therefore, in the present case, the energy release is identical with the energy balance. The energy-balance of the reaction, computed from cloud chamber studies have yielded Q-values from 2.15 to 2.66 eMV. These are too low compared to Q = 2.98 eMV obtained from mass-data. This discrepancy cannot be accounted for as being due to experimental errors. It leads one to suspect that the rather infrequent emission of α-particle in which the final nucleus $_3Li^7$ is formed in the ground state has probably been missed in the cloud chamber, the tracks recorded being those in which $_3Li^7$ is formed in one of its excited states. Consequently, the kinetic energy of the outgoing α-particle is less than its maximum value by the excitation energy of the $_3Li^7$ nucleus above the ground state. The proton-emission process has also been missed in the cloud chamber studies. If it so happens that the range of the α-particle from the first reaction and that of the proton from second reaction are accidentally almost equal, there will be serious confusion between α-tracks and proton tracks in the cloud

chamber photographs. The Q-values of the two reactions being widely different, the cloud chamber data will give misleading result.

In order to study the boron disintegration process more accurately, Maurer and Fisk (1939) adopted the second method, *viz.*, the ionisation chamber and proportional amplifier method. This method offers some obvious advantages: (1) It gives immediately the energy-balance and so makes the use of the rather inaccurate range-energy curve superfluous. (2) In the comparatively small time a large number of boron disintegrations can be observed and an energy distribution curve of the emitted particles is obtained with sufficiently low statistical error, and (3) the Q-values of α-emission and proton-emission processes being widely different, they can be easily distinguished. A careful analysis of the distribution curve led these authors to the conclusion that in the disintegration of boron by slow neutrons, both α-particles and protons are emitted, according to the reactions: ${}_5B^{10}(n, \alpha){}_3Li^7$ and ${}_5B^{10}(n, p){}_4Be^{10}$ respectively. The protons emitted in the (n, p) process do not show any discrete group-structure. The α-particles due to (n, α) process, on the other hand, show four definite groups of particles. The first of these corresponds to the formation of ${}_3Li^7$ in the ground state, while the others correspond to the three excited states ${}_3Li^7$ at 200, 410 and 640 eKV.

The accuracy of the above results is questioned by Wilson (1941). He carries out the same experiment by identical method, but does not confirm the results of Maurer and Fisk. He finds evidence for only two disintegration energies. The greater energy corresponds to the formation of ${}_3Li^7$ nucleus in the ground state and is of the value 2.99 eMV., while the smaller energy, which is released in about 93-94% of the disintegrations, is 2.57 eMV. No evidence for the (n, p) process is available. Wilson gives a detailed analysis of the factors affecting the shape of the distribution curve, and suggests that the multiple peaks in the distribution curve of Maurer and Fisk is due to distortion in their amplifier, coupled with statistical variations.

Wilson's analysis containing factors governing the form of the distribution curve obtained by this method, is of far-reaching consequence. The main considerations for such an analysis are discussed below.

FACTORS AFFECTING THE SHAPE OF THE DISTRIBUTION CURVE

(a) *Influence of the time constants of circuits on the wave-form of pulses*

If a number of pairs of ions of charge q is formed between the electrodes of an ionisation chamber of capacity C and there is a steady potential difference between the plates, the potential of the collecting plate will rise approximately linearly with time,

$$V_t = V_0(1 - e^{-t/\tau_c})$$
$$= V_0 t/\tau_c \text{ (approx.) for } t < \tau_c$$

where V_0 (maximum potential change) $= q/C$ and τ_c collecting time of the ions.

The function of the electrometer tube extends over two periods—a charging period, corresponding to the time of collection of the ions and a period of discharge necessary for the leakage. The amplitude of the pulses and their duration are determined by the time constant of the circuit assembly of the collecting electrode and the grid, which is equal to R_0C_0 where R_0 is the resistance of the whole circuit and C_0 the capacity with respect to earth. As stated by Ortner and Stetter (1929), the potential of the grid of the electrometer tube attains a value

$$V=R_0.q/\tau_c.(1-e^{-t/R_0C_0}) \quad \text{for } 0 < t \leq \tau_c)$$

while the recovery to the equilibrium potential of the grid is affected according to the equation

$$V=R_0.q/\tau_c.(1-e^{-\tau_c/R_0C_0}).\ e^{-(t-\tau_c)/R_0C_0} \quad (\text{for } t > \tau_c).$$

The time constant $K=1/R_0C_0$ thus appears to be the parameter which determines the amplitude and at the same time the duration of the pulse. The time taken to reach a maximum is that taken by the slowest ion to reach the plates. The exponential decay, depending upon K, is usually a matter of a few seconds; so it is sufficient to consider V as a constant while considering the decay of the grid voltages of the succeeding valves of the amplifier. Such an impulse is fed to the input of the first valve of the linear amplifier. The input (first) coupling circuit consists of a grid resistance R_1 in series with a grid condenser C_1. The voltage across them is V and the voltage Vg_1 across the resistance is applied to the grid of the succeeding valve. During the initial charging up period Vg_1 will increase simultaneously with V which is the varying potential on the anode of the electrometer tube. It may be shown that during the initial rise of the grid potential, Vg_1 rises according to the law

$$Vg_1=V_0.\ \tau e/\tau_c.(1-e^{-t/\tau e})$$

where $\tau e=R_1C_1$.

When V reaches its maximum value V_0 at time $t=\tau_c$, Vg_1 also attains its maximum value

$$Vg_1\ (\text{max.})=V_0\ .\ \tau e/\tau_c.(1-e^{-\tau_c/\tau e})$$
$$=V_0\ (\text{approx.}) \text{ because } \tau_c/\tau e << 1).$$

The grid potential of the succeeding valves also reach their maximum values simultaneously. In considering the subsequent behaviour of Vg_1 it is sufficient to consider V as a constant.

Thus Vg_1 decays exponentially according to the law

$$Vg_1=V_0e^{-t/\tau e}.$$

In considering the form of the impulse at later stages of amplification it will be sufficient to treat it as rising instantaneously to a certain value and then decaying according to the law given below.

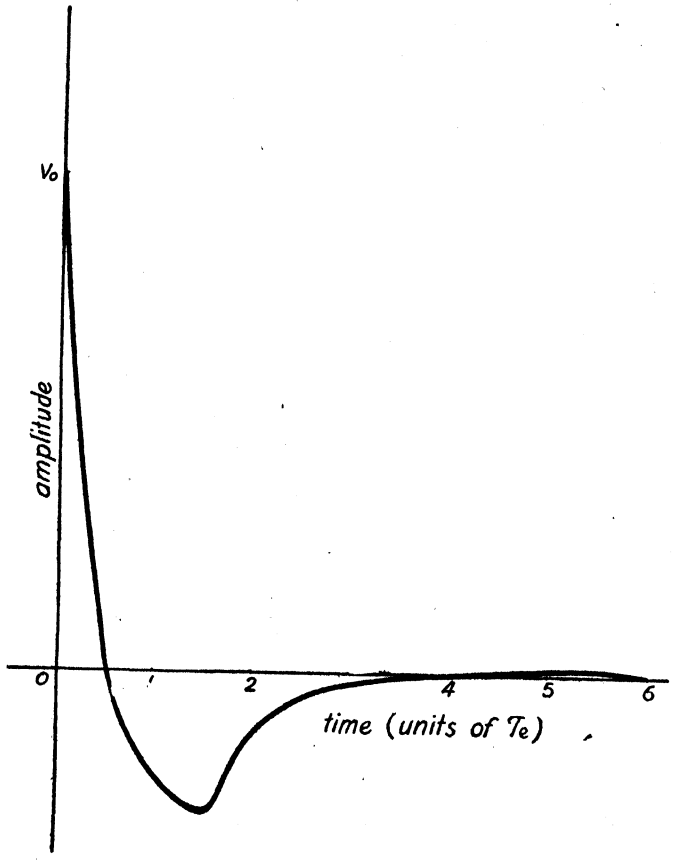
After passing through the second coupling circuit

$$Vg_2=Vg_2(\text{max})(1-t/\tau e)e^{-t/\tau e}.$$

After passing through the third coupling circuit

$$V_{g_3} = V_{g_3}(\max)(1 - 2t/\tau_e + \tfrac{1}{2}t^2/\tau_e^2)e^{-t/\tau_e}.$$

The anode current of the third valve (power valve) of the linear amplifier which actuates the oscillograph, undergoes a corresponding variation. The form of the impulse is depicted in Fig. 1. If a second pulse arrives before the original



Form of impulse after passing through three similar coupling stages

FIG. 1

one dies out, evidently the recorded size of the second pulse is different from its ideal value. This causes a broadening of the distribution curve at the skirts. A slow counting rate, depending upon the time constant of the circuit must therefore be used.

(b) Collecting time

It has been shown in the previous section that as the ions formed inside the ionisation chamber are all collected, the grid of the electrometer tube attains a value

$$V_0 = R_0 q/\tau_c.(1 - e^{-\tau_c/R_0C_0})$$
$$= q/C_0 \quad \text{when} \quad \tau_c << R_0C_0,$$

where C_0 is the capacity of the electrodes and associated circuits, and then very slowly decays exponentially to zero. Such an impulse will be distorted on passing through the intervalve couplings of the linear amplifier. We have seen that passing through the first coupling circuit,

$$V_{g_1} = V_0 \tau_e/\tau_c (1 - e^{-\tau_c/\tau_e}),$$

i.e., the maximum will be reduced to a fraction $(1 - e^{-\tau_c/\tau_e}).\tau_c/\tau_e$, of its ideal value.

For short collecting times $\tau_c << \tau_e$

$$V_{g_1} = V_0 (1 - \tfrac{1}{2}\tau_c/\tau_e),$$

i.e., the fractional reduction in voltage is $\frac{1}{2}\tau_c/\tau_e$; the effects of successive stages are additive, so that for n similar coupling stages the reduction of voltage is $n\tau_c/2\tau_e$. When disintegrations take place in a gas, τ_c will vary according to the place where the disintegration occurs, and so also with the size of kick on the oscillograph. The form of the distribution curve will consequently be broad at the peak. This broadening is greatest for long collecting times.

If therefore we wish to have a spread of less than 10% in the peak,

$$\frac{n\tau_c}{2\tau_e} < \frac{1}{10}, \text{ or, } \tau_c < \frac{2\tau_e}{10n} < \frac{\tau_e}{15}$$

for an amplifier as ours, consisting of three similar coupling stages. Thus, it is essential that the collecting time of the ions must be much smaller than the time-constant of the coupling circuits.

(*c*) *Geometry*

If disintegrations take place in the gas of an ionisation chamber, then a number of disintegration particles will strike the electrode or pass out of the effective collecting volume, and the full number of ions will not be collected. Wilson has shown that for a parallel infinite pair of plates and for an isotropic disintegration, the distribution of the lengths of the paths is represented by a back-ground of uniform height upon which is superimposed a peak of the same shape as the original but reduced in size. If the separation of the plates is d, and the length of the path of the disintegration particles is l_0, then the fraction of the total which is in the back-ground is $l_0/2d (l_0 \leq d)$. Since the shape of the peak is unaltered, it is unnecessary to use a very large separation of the plates to avoid 'edge' effects, although it is an advantage to have the diameter of the plates large compared with their separation.

From the above considerations it is obvious that the conditions for obtaining a sharp peak in the distribution curve are:

(*a*) Short collecting time
(*b*) An amplifier with coupling circuits of long time constant
(*c*) Slow counting rate.

EXPERIMENTAL ARRANGEMENT

Schematically, the experimental arrangement consists of :

(1) an ionisation chamber which collects the ions formed by the passage of the incident particle

(2) an electrometer tube which receives the charges collected and transmits them in the form of a voltage impulse

(3) a linear amplifier

(4) an oscillograph for recording the final impulse.

(1) IONISATION CHAMBER

The two essential parts of an ionisation chamber are the collecting electrode and the high voltage electrode, between which pervades the gaseous space in which the ions are formed by virtue of collision with the incident particle. In order to have a highly sensitive chamber, its capacity is made as small as possible, the variation of charge being proportional to the inverse of capacity. It is therefore advantageous to increase the depth of the chamber with electrodes of a given form inasmuch as the length of the path of the incident particle is increased and so also the number of ions produced. For this reason there is a tendency to increase the interior volume. This is, however, not always convenient. If the gas pressure is p and the separation of the plates is d, then the collecting time is proportional to pd^2 for a given collecting voltage. The edge effect is inversely porportional to pd. If we are limited, as in this case, by the total available voltage to be applied to the H. T. electrode of the ionisation chamber, then the small chamber at high pressure has the advantage over a large one at low pressures. Besides, when one works in the presence of γ rays, the number of secondary electrons produced is greater the greater the volume. This phenomenon is manifested as an increase in the back-ground hiss, thus reducing the relative sensitivity of the measurements. It is also difficult to screen a big chamber from electrical interference, as one point only is effectively earthed.

Maurer and Fisk used an ionisation chamber 30 cms. diameter and 30 cms. deep. The effective volume was enclosed between two H. T. plates 20 cms. diameter, the separation between them being also about 20 cms. The collecting electrode was situated midway between the H. T. electrodes and surrounded by an earthed guard-ring, so that the electric field between them was essentially homogeneous. A stabilised H. T. of about 8000 volts was put on the H. T. electrodes of the ionisation chamber. The chamber was filled with boron trichloride at varying pressures from 70 to 210 mm. It may be incidentally pointed out that the collecting time at the higher pressure is not lower than 1/100 sec. A weak source was kept inside an angular space situated above one of the H. T. electrodes. A narrow hole, in front of the source restricted the number of α-particles to about six per minute. A thin mica foil was also placed in front of the source so that the emerging α-rays have an energy of about the same order as the boron disintegration α-rays.

The chamber used by Wilson had plates 8 cm. diameter separated by 9 mm. and was filled with boron trichloride to a pressure of 53 cms. at 20°C. The calibration was done by means of α particles from a source of thorium active deposit, when the ionisation chamber was filled with air.

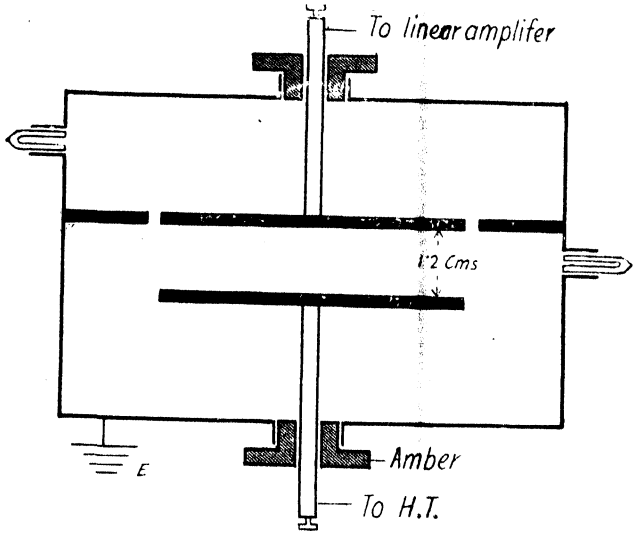


FIG. 2(*a*)

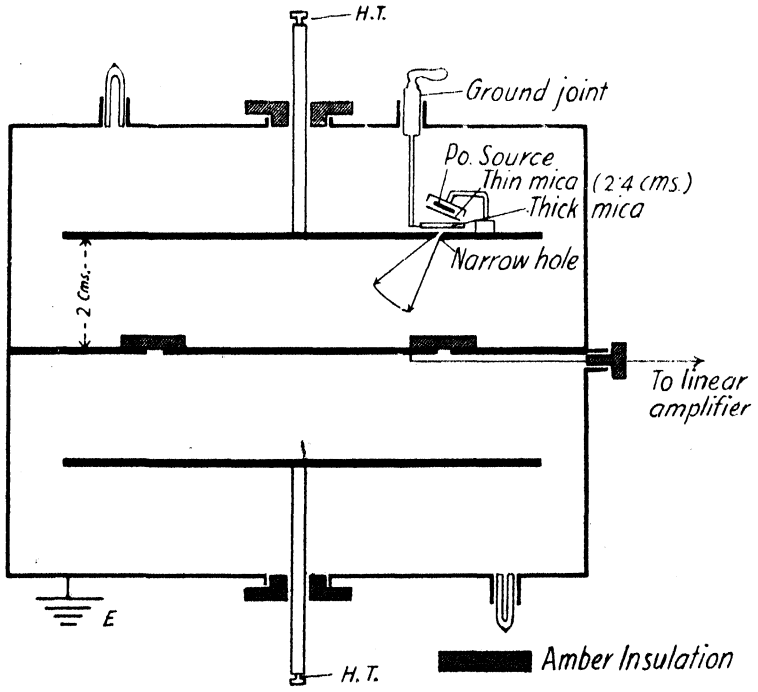


FIG. 2(*b*)

The chamber used mostly in the present experiment was essentially similar to that of Wilson. It consisted of a parallel plate condenser, the plates having

diameter of 8 cms. and separation 1.2 cms. They were housed in a cylindrical brass chamber which had "earth" potential. The leads to the plates passed through amber plugs. One of the plates was given +1800 volts, coming from a neon stabiliser. The chamber was filled with boron trichloride to a pressure of 57 cms. at 25°C. When the chamber had been filled with air, the amplifier was calibrated by means of freshly deposited Po α-rays. A second ionisation chamber, closely resembling that of Maurer and Fisk, but much reduced in size, was also used for subsidiary experiments. An essential difference in this arrangement was that the Po source was covered by a thick mica vane which completely cut off the α-rays from entering the effective volume of the chamber. By the manipulation of a ground joint from outside, the mica vane could be removed from the path of the beam when calibration was necessary. The schematic diagrams of both the chambers are given in Figs. 2(*a*) and 2(*b*).

The neutron source consisted of a glass tube containing 65 mc (Ra−Be) placed inside a lead house of 10 cms. thickness. The latter was covered on all sides by paraffin bricks of 5 cms. thickness. The source was kept at a sufficient distance from the chamber so that the maximum counting rate was about 30 per minute. The ionisation chamber was also surrounded by 5 cms. of paraffin wax. A weak (Po−Be) source was also occasionally used.

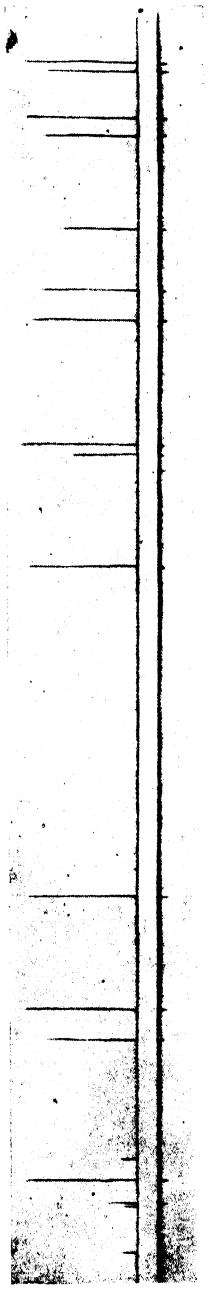
(2) FIRST STAGE: ELECTROMETER TUBE

The electrometer tube was a triode, whose highly insulated grid was directly connected to the collector electrode of the ionisation chamber; it transformed the impulses produced by the passage of α-particles across the chamber into plate current variations. In order to avoid the Johnson (1927) fluctuations in leak resistances, a floating grid was used. It was biased by itself and attained an equilibrium potential determined by the space charge of the electrometer tube. The leakage of the charges arriving at the grid was assured by the electron current itself. The tube was mounted vertically along the axis of an air-tight brass chamber, always in presence of desiccating substances. Mechanically it was supported on the shield itself by means of a very flexible rubber suspension which eliminated shocks and vibrations and also protected the valve from light.

A high positive voltage was used on the H.T. electrode of the ionisation chamber, enabling the accumulation of positive ions on the collecting electrode. This ensures a much more rapid discharge of the grid than when a negative voltage is applied, *i.e.*, when the voltage is of the same sign as the space charge of the electrons.

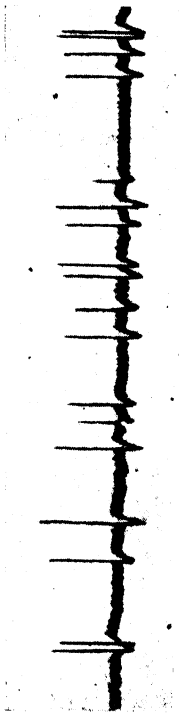
(3) SECOND STAGE: AMPLIFIER

The role of the amplifier is to amplify each pulse linearly. Using valves of very high amplification factor, one can now-a-days obtain with a set of three pentodes an amplification equal to or greater than what could be obtained previously with five or six valves; the fluctuations incumbent upon them have,



Registration of Boron—disintegration Pulses

Recording by Loop oscillograph



Recording by Cathode-ray oscilloscope

therefore, been considerably reduced. The role of amplification fell on the first two valves while the last, a power pentode, produced output pulses of sufficiently strong current to move an oscillograph.

The plate of the electrometer tube was r-c coupled to the grid of the first amplifying tube. As the method was generally used for high-speed recording of α-particles, high resolving power was necessary. Hence artificial distortion was deliberately applied for sharpening the pulses by making the time constant of the first coupling stage of the amplifier small. Wynn-Williams and Ward (1931) made the time-constant even much less with a view to amplifying only the start of the discharge and to make pulses very sharp. If it so happens, as in the case of Maurer and Fisk, that the time constant of the first coupling stage be smaller than the collecting time of the ions, the shape of the distribution curve will evidently be distorted as pointed out by Wilson. In the present experiment, the three coupling circuits had time-constants of 1/20 second each. The shielding of the elements of the amplifiers and its connection to earth together with perfect decoupling of all voltage supplies were essential precautions taken in setting up the apparatus and obtaining stable operation and a very reduced background hiss due to Shot effect. The resistances were all wire-wound and condensers were of mica.

Tests of the linear response of the amplifier and constancy of amplification were frequently made by applying a small variable alternating voltage to the ionisation chamber and measuring the corresponding amplitude of the oscillations reproduced by the oscillograph. If the amplification is proportional, the amplitude-voltage curve is a straight line. Further test was made by plotting the Bragg curve, with shallow ionisation chamber, for α-particles of known range and specific ionisation such as those emitted by Po electrochemically deposited on a silver foil, and comparing it with a standard curve.

(4) RECORDING APPARATUS

In selecting this two essential qualities need be considered: its resolving power and sensitivity. It should have sufficiently low period, less than the power of resolution of the amplifier, so that pulses which have been separately handled in the amplifier may not be superposed or confused with one another. In practice, frequencies of the order of 1000 to 5000 are enough.

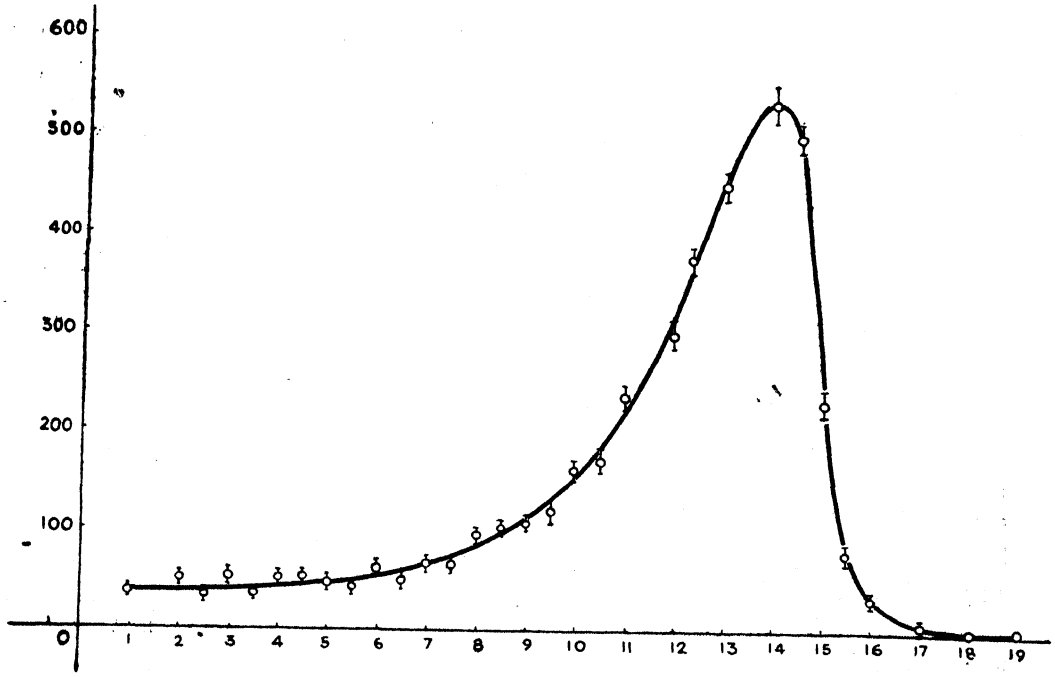
Two types of oscillograph were made and experimented with. The moving armature type was a modification of Wynn-Williams recorder. Owing to a large number of turns in the coil, it had an appreciable inductance and so caused the anode current rise exponentially with the characteristic time-constant. By using a pentode of high impedance and high mutual conductance for the output stage, this lag was appreciably reduced. The performance of the loop oscillograph was, however, very satisfactory. It consisted of a loop of ribbon running over grooves in two ivory bridges and held in tension by an ivory pulley. It carried a tiny mirror $1\frac{1}{2}$ mm. $\times \frac{1}{2}$ mm. and was situated in a

strong magnetic field. Its frequency of response was over 5000. The oscillograph was critically damped in oil. The pulses were registered on a slowly moving cine-film. A record of the pulses is shown in Plate V(*a*).

Preliminary adjustments were made by visual observations with a cathode ray oscillograph. The pulses, were usually so fast that it was difficult to get their trace on the running film. Using a higher time-constant in the last coupling stage, the pulses, though slightly distorted, were easily photographed. A record of the Cathode ray oscillograph pulses is shown in Plate V(*b*). The recording camera was specially designed for running perforated cine-films at constant speed over a widely variable range.

RESULTS

A systematic study of the shape of the distribution curve was made under different experimental conditions imposed by changing (*a*) the collecting time of ions, (*b*) the time-constant of coupling circuits and (*c*) the counting rate. The distribution curve obtained under best conditions is shown in Fig. 3. The single



Length of oscillograph deflection in mm.

FIG. 3

peak must clearly correspond to the reaction commonly observed. The energy-release is approximately 2.5 eMV. Using the ionisation chamber 2(*b*) this energy release could be conveniently compared with the pulses due to α-particles. It was found, however, that the chamber used to get contaminated within a few

days and emitted short pulses even in absence of the neutron source.* The idea of putting the calibrating source within the chamber was therefore abandoned and the distribution curve shown in Fig. 3 was obtained with chamber 2(*a*), which, incidentally, had negligible back-ground effect.

It will be seen that the second peak obtained by Wilson, corresponding to an energy release of 2.99 eMV and whose magnitude is about 6-7% of the disintegrations, has been missed in our case. Although there are oscillograph kicks representing the corresponding energy release, there is little evidence of a peak in that area in our distribution curve.

Fig. 4 shows the distribution curve under different experimental conditions. It was obtained when the Ra−Be source was kept near the ionisation chamber and therefore a much higher counting rate was employed. The curve is broadened in spite of the fact that a time-constant of 1/500 sec. was employed for the first coupling stage.

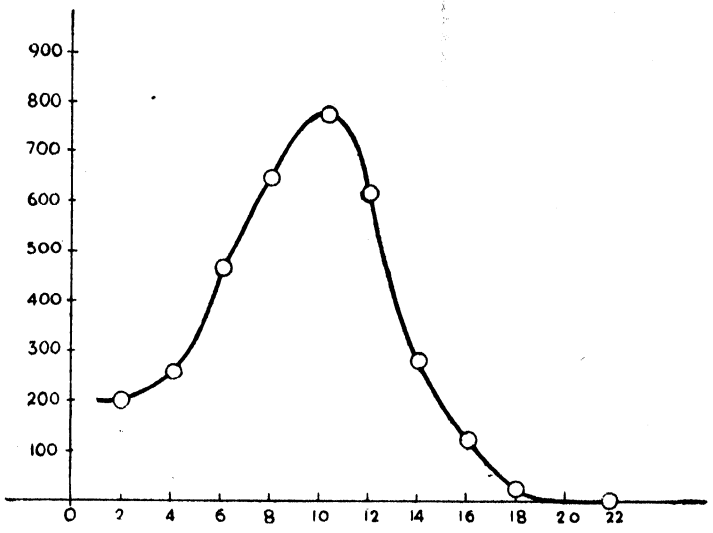


FIG. 4

DISCUSSION OF RESULTS

The experimental evidence points to a reaction energy of about 2.5 eMV. The energy calculated from mass-values gives Q=2.99 eMV. A possible explanation is that the transition to the ground state of $_3Li^7$ is forbidden and that the reaction leads to an excitation state in the $_3Li^7$ nucleus, with different angular momentum, for which the transition is allowed. The following piece of experimental evidence lends support to the above interpretation :

(1) The non-capture excitation of $_3Li^7$ by α-particles in the reaction

* It is not unlikely that the occurrence of numerous short pulses in Maurer and Fisk's arrangement, ascribed to protons, may be traced to be due to the constant presence of the calibrating source of Po within the ionisation chamber.

$_3Li^7(\alpha, \alpha+\gamma)_3Li^7$ results in γ radiation of about 0.4 – 0.6 eMV indicating such a level.

(2) Rumbaugh and Hafstad (1936) obtained two proton groups from the reaction $_3Li^6(\alpha, \gamma)_3Li^7$. The long range group probably corresponded to the formation of $_3Li^7$ in the ground state, while the short range group indicated an excitation level in $_3Li^7$ at about 0.44 eMV. The corresponding γ-ray of energy about 0.40 eMV was detected by Williams, Shepherd and Haxby (1937).

(3) According to Roberts, Heydenburg and Locher (1938) and also Maier-Leibnitz (1938) the radioactive $_4Be^7$ is accompanied by a γ-radiation of energy 0.425 eMV when it is being transformed into $_3Li^7$ by the process of β-ray decay. It is suggested that the γ-ray energy arises from a transition between the two levels in the resultant $_3Li^7$ (*e.g.*, Livingstone and Bethe (1937), Rumbaugh, Roberts and Hafstad (1938)).

(4) The $_3Li^7$ nuclei formed in the excited state in the reaction $_5B^{10}(n, \alpha)$-$_3Li^7$ will go to the ground state by the emission of a γ-ray. Fleischmann (1935) and also Kikuchi, Aoki, Husimi (1936) indicated the presence of a γ-ray. Wilson has, however, definitely proved its existence by using a boron-lined proportional counter in coincidence with an adjacent γ-ray counter. The neutron source and both counters were embedded in blocks of paraffin wax. The resolving time of the coincidence circuit was rather less than 2×10^{-6} sec. so that no coincidence between the γ-rays and neutrons emitted by the source could be recorded owing to the fact that the neutrons take a time of about 10^{-5} second to slow down to thermal velocities. The coincidences observed were therefore due to γ-rays associated in time with the α-rays from the boron disintegration. The absorption coefficient of the radiation was measured by measuring the coincidence rates when different thicknesses of lead were placed between the two counters. The energy of the observed radiation was about 0.5 eMV.

The author expresses his grateful thanks to Prof. D. M. Bose, Director, Bose Research Institute, for his kind and helpful interest in this work. His thanks are also due to Mr. S. K. Chatterjee and Dr. H. Rakshit for unfailing support.

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